

## **Impact of material heterogeneity on solute transport behavior in the unsaturated zone of the Calcaire de Beauce aquifer (France)**

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Since a few decades, the Calcaire de Beauce aquifer is contaminated with nitrate. The nitrate dynamics in the aquifer and in the surface soil are quite well understood, but its transport through the vadose zone remains largely unknown. When models fail to simulate nitrate concentrations in wells, preferential flow or physical non-equilibrium transport in soil and in the vadose zone is usually put forward to explain this failure.

To study transport processes in the vadose zone of the Calcaire de Beauce aquifer, undisturbed cores (30 cm length and 20 cm diameter) have been taken below the deepest soil horizon. At the field scale, the vadose zone is composed of powdery limestone spatially very heterogeneous, and including a variable amount of coarse elements. Two columns were selected: column "6" is made of very fine homogeneous limestone whereas column "8" is very heterogeneous with a large proportion of coarse elements.

Elution experiments have been performed on both columns. A tracer (Br- or DFBA) in a solution of 5 mM CaCl<sub>2</sub> was spread as a pulse on the top of the column with a rainfall simulator. Input flow rate was kept constant for steady state cases, or suddenly closed for flux interruption cases. Outflow was collected as a function of time for tracer concentration measurement. The collected fractions were analyzed by HPLC (High-performance liquid chromatography) with a UV detector.

Three types of experiments took place:

- For steady state experiments, three rainfall rates, respectively 4, 8, and 16 mm/h, have been used to study the occurrence of immobile water in the columns. The tracer was injected during 120 min followed by CaCl<sub>2</sub> tracer-free solution at same flow rate.
- For flux-interruption experiments, only the 4 and 8 mm/h rainfall rates were used. The tracer was injected during 120 min, input and output fluxes were then stopped and restarted seven days later with the same flow rate.
- For drainage experiments, only the 4 and 8 mm/h rainfall rates were used as well. The tracer was injected during 120 min, input flux was stopped while output flux continued to occur under the -25 cm matric head bottom boundary condition. Flux restarted seven days later with the same flow rate or another flow rate.

STANMOD was used for each BTC to estimate transport parameters assuming steady state flux. The standard CDE was suitable for column 6 steady-state experiments, but the MIM had to be used to describe properly the BTCs of column 8. In this column, the immobile water fraction represented 38 %.

Flux interruption experiments showed that the form of the BTC for Column 6 was not disturbed for the 4 and 8 mm/h input flux, whereas the form of BTC for Column 8 had significantly changed with a visible steeper increase after an interruption time compared to the corresponding steady state experiment.

This difference of behavior could be related to the difference in limestone material. The immobile water fraction was found to be significant only for columns made of heterogeneous limestone.